

Reply to “Comment on ‘Modifying the variational principle in the action-integral-functional derivation of time-dependent density functional theory’ ”

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Abstract

In a recent paper [Phys. Rev. A **77**, 062511 (2008)], it was advocated to modify the variational principle for the action-integral functional in the Runge-Gross foundation of time-dependent density-functional theory. This was criticised in a subsequent paper [Phys. Rev. A **62**, 052510 (2010)] by the present author. In a Comment [Phys. Rev. A **83**, 046501 (2011)], it is argued that the criticism is unfounded. This is a response as given to the four specific points raised in the Comment, clarifying and confirming the essence of the original criticism.

In a recent paper [1], in the following referred to as paper I, G. Vignale (GV) has advocated to modify the variational principle (VP) for the action-integral functional (AIF) in the Runge-Gross (RG) foundation of time-dependent density-functional theory (TDDFT). This was criticised in a subsequent paper [2] (paper II). In a Comment [3], GV argues that the criticism in paper II is unfounded. He tries to substantiate his case in four specific points, which will be addressed below, following a differing order here.

Time-dependent variational principle (2):

In principle, there is no disagreement here. Obviously, $\langle \delta\Psi | i\frac{\partial}{\partial t} - \hat{H} | \Psi \rangle = 0$ implies $(i\frac{\partial}{\partial t} - \hat{H})|\Psi\rangle = 0$ if the variations $|\delta\Psi\rangle$ are not restricted (exhaust the entire Hilbert space). However, as has been pointed out in paper II, this usually does not apply to a situation where one needs to resort a time-dependent VP, e.g., in establishing equations-of-motion (EOM) for approximate wave-functions. In this respect, certain formulations in paper I are misleading.

Nature of functionals (3):

In paper II (last paragraph of Sec. 4), it is not stated “that the wave function should be a functional not only of the time-dependent density itself, but also of its derivatives with respect to time.” Of course, the notion of a wave-function functional in the form $\Psi(t) = \Psi[n](t)$ is “completely general and sufficient”, since, for a given density trajectory $n(t)$ (in a time interval $[t_1, t_2]$), the first and higher time derivatives are completely determined. Nevertheless, one may ask which specific pieces of information of the density trajectory are required in order to construct the wave function *at a specific time* t according to a given functional.

However, the remark concerning the wave-function functional is only a side note. The basic issue in Sec. 4 of paper II is the representation of the AIF variation $\delta A[n]$ in terms of functional derivatives. Disregarding the phase problem for the moment, $A[n]$ is determined completely by the density trajectory $n(\mathbf{r}, t)$ in the considered time interval. However, this does not imply that the variation $\delta A[n]$ induced by a time-dependent density variation $\delta n(t)$ can be written entirely in terms of a functional derivative with respect to $\delta n(t)$, that is,

$$\delta A[n] = \int d\mathbf{r} \int_{t_1}^{t_2} dt \frac{\delta A[n]}{\delta n(\mathbf{r}, t)} \delta n(\mathbf{r}, t) \quad (1)$$

For a given density, the functional derivative is a spatially and temporally local function, $\frac{\delta A[n]}{\delta n(\mathbf{r}, t)} = f[n](\mathbf{r}, t)$. According to Eq. (1), $\delta A[n]$ would be the sum (integral) of local contributions $f[n](\mathbf{r}, t) \times \delta n(\mathbf{r}, t)$ (times $d\mathbf{r}dt$) associated with a specific point (\mathbf{r}, t) in space and time. However, it should not be taken for granted that the actual contribution to $\delta A[n]$ depends only on the static value of the density variation at time t . In view of the time-derivative in the definition of $A[n]$, one has to expect that the rate of the change of the density, that is, $d/dt\delta n(\mathbf{r}, t)$, will play a role as well.

As a pertinent example, one may consider the classical action S , briefly discussed in Sec. 2 of paper II. Obviously, the functional S is completely determined by the trajectory $q(t)$ in the considered time interval, that is, $S = S[q]$. Nevertheless, the variation δS must be written in the form

$$\delta S = \int_{t_1}^{t_2} dt (f(q, \dot{q}, t)\delta q(t) + g(q, \dot{q}, t)\delta \dot{q}(t))$$

where $f = \frac{\delta S}{\delta q(t)}$ and $g = \frac{\delta S}{\delta \dot{q}(t)}$ are the (temporal) functional derivatives of S with respect to $q(t)$ and $\dot{q}(t)$, respectively. (At a given *point in time*, t , the velocity $\delta \dot{q}(t)$ cannot be inferred from $\delta q(t)$, but must be supplied independently.)

It should be noted that the representation of $\delta A[n]$ according to Eq. (1) was introduced in the famous 1984 paper by Runge and Gross [4] without discussion, and has been seen as quasi self-evident ever since. Actually, it must be seen as the second fatal error in the original TDDFT foundation (the first one being the indefiniteness of the action integral functional).

The “loophole” (4):

The “loophole” in the argument for modifying the VP in paper I is not “constructed,” but rather obvious: for the exact density (not to be dismissed as “certain particular exact densities”) the boundary term $\langle \Psi[n](t_2) | \delta \Psi[n, \delta n](t_2) \rangle$ could vanish due to orthogonality of $|\Psi[n](t_2)\rangle$ and $|\delta \Psi[n, \delta n](t_2)\rangle$. As long as this possibility cannot be excluded, the case for a modified VP is not stringent.

Definition of the action functional (1):

This is the crucial issue. Here GV writes: “This [the finding that the RG action-integral functional is ill-defined] is false, because multiplying the wave function by a time-dependent

phase factor $e^{-i\alpha[n](t)}$, where $\alpha[n](t)$ is an arbitrary functional of the density, n , and a function of time, t , amounts to adding to the Lagrangian the total time derivative $\frac{d\alpha[n](t)}{dt}$. It is generally the case ... that the Lagrangian is defined up to an arbitrary time derivative of a function of the coordinates and time: it is well known that this 'gauge freedom' does not affect the *variation* of the action and therefore leaves the equations of motion unchanged."

There are two points to be made here:

- Firstly, the problem with the phase function is that $\alpha(t)$ is completely arbitrary (apart from the possibility to fix it at the initial time t_1) and, in fact, cannot be presented as being a *functional*, $\alpha[n](t)$, of the density. The tacit assumption that the phase factor is of the form $e^{-i\alpha[n](t)}$ with an arbitrary but otherwise well-defined functional $\alpha[n]$ amounts, logically speaking, to a *petitio principii*. I shall come back to this point below.

- Secondly, an indefiniteness of the Lagrangian with respect to a total time derivative $\frac{d\alpha(t)}{dt}$ is permissible only if the variation of the boundary term $\alpha(t_2) - \alpha(t_1)$ in the action integral vanishes. However, this does not apply to the RG AIF and the original VP, since the variation of the phase, $\delta\alpha(t_2)$, cannot be assumed to vanish at the boundary t_2 of the time interval. (In fact, the (density induced) variation of the wave function itself does not vanish here, i.e. $\delta\Psi(t_2) \neq 0$, as GV rightly observes.) So there should be agreement that, at least in conjunction with the original VP, the RG AIF is ill-defined.

Now let us consider the modified VP. GV claims that the new VP (Eq. 1 in the Comment, referred to in the following as Eq. C-1) is "completely unaffected by the arbitrary phase". This is demonstrated by a small derivation, of which only the resulting Eq. (C-2) is given in C. (For a better understanding of this argument a detailed derivation of Eq. (C-2) is presented in the appendix below.) The problem with this demonstration is the unwarranted use of the arbitrary phase function as a functional of the density, $\alpha = \alpha[n](t)$. The derivation of Eq. (C-2) depends manifestly on the possibility to expand the phase function for the varied density, $n(t) + \delta[n](t)$, according to $\alpha[n + \delta n](t) = \alpha[n](t) + \delta\alpha[n](t)$. However, the actual situation is beyond remedy. Even if one assumes that the wave function $\Psi[n](t)$ for a specific $n(t)$ comes with a defined time-dependent phase function $\alpha(t)$, any phase function $\tilde{\alpha}(t)$ (with $\tilde{\alpha}(t_1) = 0$) would be permissible for the varied wave function $\Psi[n + \delta n](t)$, so that something to the effect of a variation of the phase function, $\delta\alpha(t)$, cannot even be properly defined. This means that the indefiniteness of the AIF is not at all cured by the new VP.

A final remark concerning gauge transformations: Of course, the *physics* must be

invariant with respect to gauge transformations, but what we are dealing with here is primarily a *mathematical* issue, namely an attempt to replace the time-dependent Schrödinger equation by an equivalent density-based EOM. Mathematically, it is manifest that a given density determines the corresponding wave function only up to an arbitrary time-dependent phase function, and this has mathematical implications, which should not be dismissed by referring to a physical principle.

In view of the response given above to the four objections raised in the Comment, there is no justification for the statement that “Schirmer’s critique of my paper is invalid, and my reformulation of the variational principle and the resolution of the causality paradox ... stand in their pristine form.” On the contrary, the essence of our criticism has not been rebutted:

- (i) The original RG AIF is ill-defined due to the arbitrary purely time-dependent phase, and this problem is not eliminated in the original variational procedure.
- (ii) The case for a modification of the VP is not rigorous, nor is the phase problem in the definition of the AIF overcome by the modified VP.
- (iii) Irrespective of the phase problem, the variation of the AIF cannot be expressed entirely in terms of a functional derivative with respect to the density. It must be expected that functional derivatives with respect to the first (and possibly second) time derivatives of the density come into play.

Appendix: Derivation of Eq. (C-2)

The idea is to compare the VP for a phase-augmented wave function functional (WFF), $\tilde{\Psi}[n](t) = e^{-i\alpha[n](t)}\Psi[n](t)$ with that for the original one, $\Psi[n](t)$. The left-hand side of Eq. (C-2) can readily be established, $\delta A[n]$ denoting the variation of the WFF without the additional phase. For the evaluation of the right-hand side of Eq. (C-1), the starting point is

$$\delta\tilde{\Psi}[n](t) = e^{-i\alpha[n+\delta n](t)}\Psi[n+\delta n](t) - e^{-i\alpha[n](t)}\Psi[n](t) \quad (2)$$

Using the expansions

$$\Psi[n+\delta n](t) = \Psi[n](t) + \delta\Psi[n](t) \quad (3)$$

$$\alpha[n+\delta n](t) = \alpha[n](t) + \delta\alpha[n](t) \quad (4)$$

gives

$$\delta\tilde{\Psi}[n](t) = e^{-i\alpha[n](t)}[e^{-i\delta\alpha[n](t)}(\Psi[n](t) + \delta\Psi[n](t)) - \Psi[n](t)] \quad (5)$$

Now, the scalar product on the right-hand side of Eq. (C-1) can be evaluated (for t_2) according to

$$\langle\tilde{\Psi}[n](t_2)|\delta\tilde{\Psi}[n](t_2)\rangle = e^{-i\delta\alpha[n](t_2)} + e^{-i\delta\alpha[n](t_2)}\langle\Psi[n](t_2)|\delta\Psi[n](t_2)\rangle - 1 \quad (6)$$

Finally, using the expansion of the exponential function,

$$e^{-i\delta\alpha[n](t)} = 1 - i\delta\alpha[n](t) \quad (7)$$

the right-hand side of Eq. (C-1) becomes (through first order in the variations)

$$i\langle\tilde{\Psi}[n](t_2)|\delta\tilde{\Psi}[n](t_2)\rangle = \delta\alpha[n](t_2) + i\langle\Psi[n](t_2)|\delta\Psi[n](t_2)\rangle \quad (8)$$

thus completing the proof of Eq. (C-2). The variation of the phase drops out of Eq. (C-2), implying that the modified VP is not affected by adding a purely time-dependent phase factor to the wave function. While there is nothing wrong with the calculation itself, the problem resides in the tacitly assumed premise that the arbitrary TD phase can be treated as a functional $\alpha[n](t)$ of the density.

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- [1] G. Vignale, Phys. Rev. A **77**, 062511 (2008).
 - [2] J. Schirmer, Phys. Rev. A **82**, 052510 (2010).
 - [3] G. Vignale, Phys. Rev. A **83**, 046501 (2011).
 - [4] E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).